

(iii) treating the stream recovered in step (ii) to contain less than 50 wppm nitrogen in the form of organic nitrogen compounds to obtain a treated stream, and

(iv) recycling at least part of the treated stream of step (iii) to the epoxidation reaction step (i),

wherein one or more nitrogen containing compounds are introduced at some stage of the process.

It is known in the prior art that olefins can be converted by reaction together with hydrogen peroxide to form epoxides when a titanium containing zeolite catalyst is used. This is known from the European patent document 100118. The reaction is carried out in the presence of polar solvent which functions to dissolve both the olefin and the hydrogen peroxide in a reaction mixture.

It is also known to introduce nitrogen containing compounds to an olefin-hydrogen peroxide epoxidation reaction to improve the propylene oxide selectivity. The prior art discloses a number of nitrogen containing compounds that can be used for this purpose such as shown in European patent A 230949.

When the epoxidation process is performed on an industrial scale, the polar solvent used in the epoxidation reaction has to be recovered in the work-up of the reaction mixture and recycled to the epoxidation reaction system for economic reasons. However, when this solvent is recycled, impurities contained in the recycled solvent may accumulate in the recycle up to undesirable levels.

Applicants have discovered that the recycled solvent may also contain substances which poison the epoxidation catalysts, which leads to lower catalytic activity and a more rapid

deactivation of the epoxidation catalyst. See, page 5, para. 0017. Indeed, applicants have observed the occurrence of such catalyst poisoning when one or more of the nitrogen containing compounds are introduced in some stage of the process. Applicants have also observed that the known methods for recovering and recycling the solvent have not overcome the problem of catalyst poisoning.

Thus, the state of the art before applicants' invention was that (1) a polar solvent was desired for the reaction of the olefin and hydrogen peroxide, (2) nitrogen compounds were desired to be present to improve the selectivity of the reaction, and (3) the solvent had to be recycled in order for the process to be economical on a commercial scale.

Applicants have observed that when the recovered polar solvent is recycled to the epoxidation step, this leads to a reduced activity of the epoxidation catalyst and a more rapid catalyst deactivation when one or more nitrogen containing compounds are introduced at some stage of the epoxidation process utilizing a titanium containing zeolite catalyst.

It was then surprisingly discovered by applicants that the epoxidation catalysts can be prevented from suffering a serious reduction in activity by treating the recovered stream containing the polar solvent in such a way that it contains less than 50 wppm nitrogen in the form of organic nitrogen compounds before recycling the polar solvent to the epoxidation step. As explained in the application, the abbreviation "wppm" means weight parts per million.

In a more preferred aspect of the invention, the recovered polar solvent stream is treated to contain less than 30 wppm nitrogen in the form of organic nitrogen compounds and still more preferably less than 15 wppm and most preferably less than 10 wppm nitrogen.

Thus, applicants' invention resides in substantially reducing the amount of nitrogen in the form of organic nitrogen compounds in the polar solvent to be recycled before the polar solvent is recycled to the epoxidation step. This feature and the present invention as a whole is neither described nor even remotely suggested by the combination of references identified in the Official Action of October 31, 2005. Indeed, the data in the application on page 20 in table 1 clearly shows that the presence of organic nitrogen compounds in the amount of more than 50 wppm (example 1) results in a much lower hydrogen peroxide conversation and a greatly reduced yield of propene oxide.

Thus, in summary, while the skilled person in the art was aware that in the process for the catalytic epoxidation of olefins in the presence of a titanium containing zeolite catalysts, the introduction of nitrogen containing compounds was desirable to obtain certain improved results, the prior art was unaware of the problem of catalyst poisoning.

Turning now to the discussion in the Official Action concerning the prior art, the Official Action notes that the European patent document A 100118 "teach using hydrogen peroxide and an olefin in the presence of a titanium catalyst to produce an epoxide." This is essentially correct and comports with applicants' discussion of this reference found in the application on page 1 in paragraph 0003.

The statement in the Official Action also acknowledges that European patent document A 230949 teaches "nitrogen containing compounds added to epoxidation reaction to improve the epoxide selectivity". This statement also comports with applicants' disclosure in the application on page 1, paragraph 0006.

The Official Action relies on the *Paparatto* reference for a teaching of “separating the solvent from the epoxidation mixture, then removing (eliminating) the nitrogenated compounds present, and recycling the solvent into step (1) of the epoxidation process.”

The Official Action continues (page 3) by stating “The difference is that *Paparatto* teach ‘elimination of the nitrogenated compounds present’ while applicants teach -- limiting the amount of nitrogenated compounds to less than 50 wppm”.

Applicants disagree with the conclusion in the Official Action that *Paparatto* teaches recycling of solvent which has the nitrogen content “modified to increase epoxide selectivity” and will show below that this conclusion has no logical basis. Applicants submit that one of ordinary skill in the art would not have been motivated to modify the process of either of the European patents to arrive at the claimed process.<sup>1</sup>

*Paparatto*, WO 02/14299, discloses an integrated process for making olefin oxides where, in a first step, an alcoholic solution of hydrogen peroxide is prepared by reacting hydrogen and oxygen in presence of a palladium platinum catalyst. Thereafter, the hydroperoxide solution is reacted with olefin and the solvent recovered from the reaction product is treated to remove nitrogen-containing compounds. Thereafter, the purified solvent is recycled into step 1, i.e., the preparation of the hydrogen peroxide solution. That is a critical distinction with respect to the process of the present invention because the present invention does not involve a recycle to the step of hydrogen peroxide preparation.

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<sup>1</sup> It is noted the Office Action refers to modifying the “chemical process” to achieve the same result; see pg. 3, lines 5-6. Presumably, the Office Action intended to refer to the processes of either or both of the European patents since the “claimed process” is not part of the prior art.

In the Office Action, *Paparatto* is relied on to show recycling of the solvent into the epoxidation step. However, *Paparatto* does not recycle solvent to the epoxidation step, but instead to a preliminary reaction of the hydrogen peroxide preparation step. This is not the same as recycling to the epoxidation step. Present claim 1 clearly specifies the purified solvent is recycled into the epoxidation step.

**It is, therefore, apparent that no prior art has been cited to show recycling a solvent with an organic nitrogen content of less than 50 wppm to the epoxidation step.**

Furthermore, the statement by the Examiner on page 3, second paragraph, of the Office Action arguing that the *Paparatto et al.* application teaches that reduction of the nitrogen content would lead to an improved epoxide selectivity is clearly wrong and contrary to the disclosure of the *Paparatto* reference.

On page 19, lines 1 to 4, of the *Paparatto* reference it is clearly stated that the function of the acid treatment is to completely separate the nitrogenated basic products present in the stream, which could influence the performance of the catalyst used for the synthesis of hydrogen peroxide. Consequently, according to the teaching of the *Paparatto* reference, a removal of nitrogenated basic compound is only necessary **if** an integrated process comprising the step of direct preparation of hydrogen peroxide in presence of noble metal catalysts is used. In the *Paparatto, et al.* reference, the problem of catalyst poisoning of recycled solvent is only addressed with respect to the noble metal catalyst used for making the hydrogen peroxide solution, but there is no recognition in the art that organic ammonium compound present in the recycled solvent may cause deactivation of the titanium silicalite catalyst in the epoxidation step. Thus, there is no motivation at all for a person skilled in the art to introduce an additional step of

removing nitrogenated bases in a process as presently claimed wherein no direct preparation of hydrogen peroxide is used as step of an integrated process.

Furthermore, applicants point out that, according to the teaching of the *Paparatto* reference, as defined in claim 1 in his PCT published application, the epoxidation of olefins is conducted in presence of a buffering agent. On page 22, lines 2 to 6, of the *Paparatto* reference it is explicitly stated that the buffering agent is selected from ammonia, ammonium acetate, ammonium formate or a system consisting of a nitrogenated base and one of its salts with an organic or inorganic acid as described in Italian patent application MI 99A/001658, which is equivalent to U.S. patent 6,300,506. U.S. patent 6,300,506 discloses in col. 2, lines 23 to 50, that an alkylamine can be used as the nitrogenated base. Consequently, it is the explicit teaching of the *Paparatto* reference to have a nitrogen-containing base like alkylamine present in the epoxidation step.

Moreover, in *Paparatto, et al.*, a preferred embodiment according to Claim 3 and the specification on pg. 17, lines 12-24 the filtered liquid product leaving the epoxidation reaction unit is fed to distillation wherein in the flash column the product is separated into a head product and a bottom product consisting essentially of solvent non-reacted hydrogen peroxide, water and reaction by-products. This bottom product, in case the hydrogen peroxide concentration fed to the epoxidation process is too high, is recycled without any further treatment to the epoxidation step. Due to the presence of ammonium compounds as possible buffers in the reaction step, these ammonium compounds in the flash distillation will concentrate in the bottom product and will be recycled without any further treatment into the epoxidation step of the process. Thus, contrary to the allegation in the Office Action, *Paparatto, et al.* explicitly teaches to recycle the

alcoholic solvents separated from the epoxidation product directly without any further treatment into the epoxidation process, thus also recycling any ammonium compounds directly into the epoxidation step. This further shown that *Paparatto, et al.* lead the skilled worker into a totally different direction compared to the present invention.

Still further, with respect to the removal of nitrogen containing compounds from solvents that are recycled to the direct epoxidation step for preparing hydrogen peroxide, *Paparatto, et al.* does not discriminate between organic and inorganic nitrogen compounds. Furthermore, *Paparatto, et al.* teaches that to the hydrogen peroxide containing solution in the epoxidation reaction step, a buffer compound is added which can contain a nitrogen containing compound.

But according to the present invention, it is important that the amount of organic nitrogen compounds be reduced to be less than 50 weight ppm in the recycled solvent, whereas inorganic nitrogen compounds like ammonia do not create any problem and can be tolerated also in considerably higher amounts during the epoxidation step.

Taking also into account the teaching of EP-A-230949 disclosing that the selectivity to the desired epoxide in the epoxidation reaction of hydrogen peroxide with olefin in presence of a titanium silicalite catalyst is improved if nitrogen-containing bases are present, all the references, including the *Paparatto* reference, teach that the presence of nitrogen-containing bases in the epoxidation process is beneficial for the epoxidation reaction. This is clearly contrary to the teaching of the present invention wherein it was discovered, as explained in some detail in the present application, that nitrogen-containing organic compounds should be removed from the epoxidation reaction system or restricted to less than 50 wppm.

The cited prior art does not suggest at all that nitrogen containing compounds may adversely effect the performance of the epoxidation catalyst, but even more, no hint can be derived from the prior art that organic ammonium compounds differ from inorganic compounds, like ammonia, in their detrimental effect on the catalyst performance.

Consequently, a person skilled in the art would not find any motivation – neither in one or both European references nor in the *Paparatto* reference – to remove organic nitrogen-containing compounds from a process stream that is recycled into the epoxidation step because all three references teach that nitrogen-containing bases should be present during the epoxidation. Thus, to remove a compound that, according to the teaching of these references, should be present under any circumstance during the reaction step would be contrary to the clear teaching of these prior art references.

The references, either individually or collectively, fail to provide any reason or suggestion that a benefit or advantage could be obtained by reducing the amount of organic nitrogen compound content of the recycled solvent to the epoxidation reaction as defined in the claims.

In the absence of any reason or suggestion in the references of an advantage or benefit, there is no motivation for a person skilled in the art to modify or change the processes described in the two European references.

Absent a motivation, the combination of references of record herein cannot establish *prima facie* obviousness. See, MPEP, § 2142 which says, in part:

To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally



available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. Second, there must be a reasonable expectation of success. Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations. The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art, and not based on applicant's disclosure. *In re Vaeck*, 947 F.2d 488, 20 USPQ2d 1438 (Fed. Cir. 1991).

The rejection of Claims 1 to 52 under 35 U.S.C. § 103(a) is deemed improper and should be withdrawn.

Favorable action at the Examiner's earliest convenience is respectfully requested.

Respectfully submitted,

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